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(21) International Application Number: PCT/NL99/00331 (22) International Filing Date: 27 May 1999 (27.05.99) (30) Priority Data: 1009288 29 May 1998 (29.05.98) NL (71) Applicant (for all designated States except US): DSM N.V. [NL/NL]; Het Overloon 1, NL-6411 TE Heerlen (NL). (72) Inventors; and (75) Inventors/Applicants (for US only): DIJKSTRA, Krijn [NL/NL]; Straatburglaan 54, NL-6137 JB Sittard (NL). GJUSMAN, Pieter [NL/NL]; Felix Ruttenlaan 63, NL-6132 CW Sittard (NL). (74) Agent: ALFENAAR, Marinus; Octrooibureau DSM, P.O. Box 9, NL-6160 MA Geleen (NL).		(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW. ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i>
(54) Title: UV STABLE POLYETHERESTER COPOLYMER COMPOSITION AND FILM THEREFROM		
(57) Abstract <p>The invention relates to a polyetherester copolymer composition having a strongly improved UV stability, which is eminently suitable for use in film. The polyetherester copolymer composition according to the invention contains 0.1–10 wt.% activated carbon and 0.05–3 wt.% of a hindered amine light stabilizer (HALS). Preferably, the contents are 0.5–3 and 0.2–1 wt.%, respectively. At a film thickness of 100 µm an increase in the life on the order of a factor 2, relative to the effect of the sum of the individual components, appears to be possible. Preferably, the molecular weight of the HALS is higher than 1500.</p>		

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UV STABLE POLYETHERESTER COPOLYMER
COMPOSITION AND FILM THEREFROM

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The invention relates to elastomer copolyetherester film having improved resistance to ageing under the influence of UV light. Elastomer copolyetherester film is increasingly being used as a moisture regulating top layer in roof coverings and other building applications. During the building phase the film is often exposed to direct sunlight for a longer period of time before the roof tiles are placed. Especially in mountainous regions the irradiation by UV light can be very intensive. UV irradiation causes the mechanical properties of the unprotected film to deteriorate very rapidly, and as a result the film may very easily be damaged, for example by falling tools. In more extreme cases it is no longer possible to tread on the roof covering without damaging it. In addition, the moisture regulating properties are adversely affected.

From Encyclopaedia of Polymer Science and Engineering Vol. 12, p. 102 (1988), John Wiley and Sons, Inc., it is known that copolyetheresters can effectively be protected against UV light in outdoor applications by the addition of 0.5-3 wt.% activated carbon. Vol. 15, pp. 563-564 (1989) of the same Encyclopaedia further discloses that good dispersion of the activated carbon is essential and the particle size should be between 15 and 25 nm. The claims of EP-0783016-A2 again repeat these findings, on account of which copolyetherester film would have a good UV stability and improved IR remission. The present inventors, however, have not been able to find any

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experimental basis in EP-0783016-A2 for said advantages or for any special effect. On the contrary, the UV resistance of carbon black containing copolyetherester film, in particular of the membranes disclosed in claim 5 14 of said patent, with a thickness of 5-30 μm , proves to differ only slightly from that of non-stabilized membranes.

The UV-stabilized copolyetherester films according to the state of the art appear to be wholly 10 incapable of meeting the rigid conditions that are imposed on films in particular in the building world.

The aim of the invention is therefore a copolyetherester composition which, also when used in very thin film, has a considerably improved resistance 15 to UV irradiation so as to enable application inter alia as a moisture regulating top layer in roof covering.

The inventors have achieved this aim with a copolyetherester composition containing a stabilizer 20 combination of 0.1-10 wt.% activated carbon and 0.1-3 wt.% of a hindered amine light stabilizer (HALS). The use of HALS in a copolyetherester composition to improve the stability has been known for a long time, its use being disclosed inter alia in JP-52-044869-A. 25 To improve its activity, it is known to apply combinations of HALS with other UV stabilizers. JP-60-015455-A, US-4,524,165-A and JP-02-283754 describe the combination with a triazole and JP-04-337.349-A the combination with a thio-ether. The inventors have 30 tested these combinations and some other combinations of UV stabilizers, but none of these yielded enough stability for the use in film.

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It is highly surprising that the combination with activated carbon, conversely, yields a very substantial improvement of the resistance to UV radiation, to such an extent that even very thin
5 polyetherester film can be exposed to direct UV light for a longer period of time.

The copolyetheresters of the composition comprise copolyetheresters derived from polyether glycols with an average molecular weight of 600-6000,
10 glycols, preferably alkylene glycols, for example ethylene or butylene glycol, and dicarboxylic acids, for example aromatic dicarboxylic acids, preferably terephthalic acid and naphthalenic dicarboxylic acid, cycloaliphatic dicarboxylic acids, for example
15 cyclohexane dicarboxylic acid and aliphatic dicarboxylic acids, for example adipic acid.

Examples and preparation of such copolyetheresters are described in, inter alia, Thermoplastic Elastomers, 2nd Ed., Chapter 8, Carl
20 Hanser Verlag (1996) ISBN 1-56990-205-4, Handbook of Thermoplastics, ed. O. Otabisi, Chapter 17, Marcel Dekker Inc., New York 1997, ISBN 0-8247-9797-3 and the Encyclopaedia of Polymer Science and Engineering, Vol. 12, pp. 75-117 and the references cited therein. A
25 large number of copolyetheresters are commercially available.

The ratio between the polyester units derived from the polyether glycol and those derived from the alkylene glycol, i.e. the ratio between the
30 soft and hard segments of the copolyetherester may vary within a broad range and is determined mainly by the application of the polyetherester copolymer. In general this ratio will be chosen so that the Shore D hardness of the polymer is between 25 and 80.

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The moisture-regulating capacity of the membranes obtained from the polyetherester copolymer is varied as needed through variation of the thickness of the membrane or by varying the C:O ratio in the polyalkylene oxide glycols in the polyetherester copolymer. In general the ratio is between 2 and 4.3, a low ratio generally leading to an increase in the water vapour permeability.

Activated carbon is in this context understood to be a specific form of carbon consisting of very fine primary particles combined into primary aggregates, which are not broken under normal dispersing conditions. These primary aggregates in turn generally combine in the form of agglomerates. Characteristic of this activated carbon is the large surface area that is optically available. This surface area is determined mainly by the size of the primary particles and their degree of packing in the primary aggregates. Activated carbon can be obtained in different manners and is commercially available in different forms and under various tradenames. Preferably, carbon black is used. It is preferred that under the conditions of preparation of the composition all agglomerates are broken down into primary aggregates, which should form a fine dispersion in the composition. The diameter of the primary particles may vary within a broad range, for example between 10 and 100 nm. Preferably, the average particle size is chosen to be between about 15 and 40 nm. When the particles are smaller, the radiation absorbing capacity decreases strongly as the radiation can bend around the particle. When the particles are bigger, the optically available surface area decreases too strongly, and the diameter of the primary aggregates may sometimes be too large as

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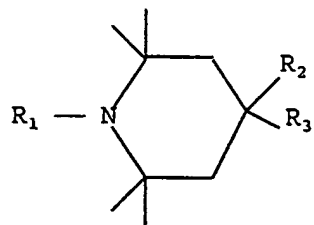
a result of which processing into thin films presents problems.

The activated carbon content of the composition may in principle vary within a broad range, for example between 0.1 and 10 wt.%. Preferably, the content is limited to between 0.5 and 5 wt.%, even more preferably between 0.5 and 3 wt.%, as lower concentrations have relatively little effect and at concentrations in excess of 3 wt.% only little more UV radiation is absorbed. The useful upper limit is generally the content at which the surface layer of the composition just ceases to be transparent. For very thin film, therefore, a higher carbon content is desirable than for thicker film. Higher carbon contents, incidentally, generally have an adverse effect on the mechanical properties, such as elongation at break.

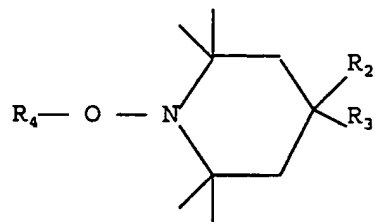
The hindered amine light stabilizer (HALS) of the composition according to the invention is a UV light stabilizer for copolyetherester compositions that in itself is often used by ones skilled in the art. Very well-known in the professional world is the HALS disclosed in the European patent EP-000389-B1, bis-(1,2,2,6,6-penta-methyl-piperidyl)-(3',5'-di-tert-butyl-4'-hydroxy-benzyl)-butyl-malonate, available under the tradename Tinuvin® 144 from Ciba-Geigy.

A hindered amine light stabilizer is understood to mean compounds of the following general formulas:

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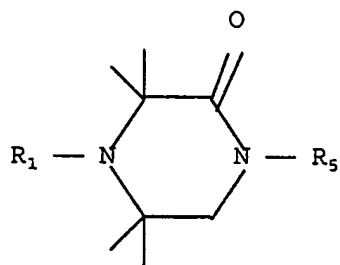


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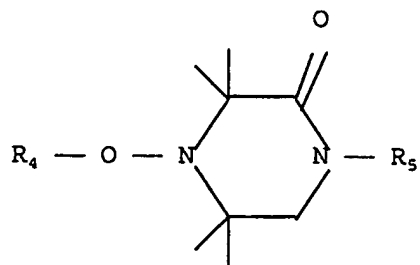
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and combinations thereof.

In these formulas, R_1 up to and including R_5 are independent substituents. Examples of suitable substituents are hydrogen, ether groups, ester groups, amine groups, amide groups, alkyl groups, alkenyl groups, alkynyl groups, aralkyl groups, cycloalkyl groups and aryl groups, in which the substituents in turn may contain functional groups; examples of functional groups are alcohols, ketones, anhydrides, imines, siloxanes, ethers, carboxyl groups, aldehydes, esters, amides, imides, amines, nitriles, ethers, urethanes and any combination thereof. A hindered amine light stabilizer may also form part of a polymer.

Preferably, as HALS compound a compound is chosen that is derived from a substituted piperidine compound, in particular any compound derived from an alkyl-substituted piperidyl, piperidinyl or piperazinone compound, and substituted alkoxypiperidinyl compounds. Examples of such compounds are:

- 2,2,6,6-tetramethyl-4-piperidone;
- 2,2,6,6-tetramethyl-4-piperidinol;
- bis-(1,2,2,6,6-pentamethyl piperidyl)-(3',5'-di-tert-butyl-4'-hydroxybenzyl)-butylmalonate;
- di-(2,2,6,6-tetramethyl-4-piperidyl)-sebacate (Tinuvin® 770) ;
- oligomer of N-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-piperidinol and succinic acid (Tinuvin® 622);

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- oligomer of cyanuric acid and N,N-di(2,2,6,6-tetramethyl-4-piperidyl)-hexamethylene diamine;
- bis-(2,2,6,6-tetramethyl-4-piperidinyl)-succinate;
- bis-(1-octyloxy-2,2,6,6-tetramethyl-4-piperidinyl)-
- 5 sebacate (Tinuvin® 123);
- bis-(1,2,2,6,6-pentamethyl-4-piperidinyl)-sebacate (Tinuvin® 765);
- tetrakis-(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butane-tetracarboxylate;
- 10 - N,N'-bis-(2,2,6,6-tetramethyl-4-piperidyl)-hexane-1,6-diamine (Chimasorb® T5);
- N-butyl-2,2,6,6-tetramethyl-4-piperidinamine;
- 2,2'-[(2,2,6,6-tetramethyl-piperidinyl)-imino]-bis-[ethanol];
- 15 - poly((6-morpholine-S-triazine-2,4-diyl)(2,2,6,6-tetramethyl-4-piperidinyl)-imino)hexamethylene-(2,2,6,6-tetramethyl-4-piperidinyl)-imino) (Cyasorb® UV 3346);
- 5-(2,2,6,6-tetramethyl-4-piperidinyl)-2-cyclo-undecyl-oxazole) (Hostavin® N20);
- 20 - 1,1'-(1,2-ethane-di-yl)-bis-(3,3',5,5'-tetramethyl-piperazinone);
- 8-acetyl-3-dothecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro(4,5)decane-2,4-dione;
- polymethylpropyl-3-oxy-[4(2,2,6,6-tetramethyl)-
- 25 piperidinyl)-siloxane (Uvasil® 299);
- 1,2,3,4-butane-tetracarboxylic acid-1,2,3-tris(1,2,2,6,6-pentamethyl-4-piperidinyl)-4-tridecylester;

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- copolymer of alpha-methylstyrene-N-(2,2,6,6-tetramethyl-4-piperidiny1)maleimide and N-stearyl maleimide;
- 1,2,3,4-butanetetracarboxylic acid, polymer with
5 beta,beta,beta',beta'-tetramethyl-2,4,8,10-tetraoxaspiro[5.5]undecane-3,9-diethanol, 1,2,2,6,6-pentamethyl-4-piperidiny1 ester (Mark® LA63);
- 2,4,8,10-tetraoxaspiro[5.5]undecane-3,9-diethanol,beta,beta,beta',beta'-tetramethyl-polymer
10 with 1,2,3,4-butanetetracarboxylic acid, 2,2,6,6-tetramethyl-4-piperidiny1 ester (Mark® LA68);
- D-glucitol, 1,3:2,4-bis-O-(2,2,6,6-tetramethyl-4-piperidiny1idene)- (HALS 7);
- oligomer of 7-oxa-3,20-diazadispiro[5.1.11.2]-
15 heneicosan-21-one,2,2,4,4-tetramethyl-20-(oxiranylmethyl)- (Hostavin® N30);
- propanedioic acid, [(4-methoxyphenyl)methylene]-,bis(1,2,2,6,6-pentamethyl-4-piperidiny1) ester (Sanduvor® PR 31);
- 20 - formamide, N,N'-1,6-hexanediylbis[N-(2,2,6,6-tetramethyl-4-piperidiny1 (Uvinul® 4050H).
- 1,3,5-triazine-2,4,6-triamine, N,N'''-[1,2-ethanediylbis [[4,6-bis[butyl(1,2,2,6,6-pentamethyl-4-piperidiny1)amino]-1,3,5-triazine-2-
25 yl]imino]-3,1-propanediyl]]-bis[N',N''-dibutyl-N',N''-bis(1,2,2,6,6-pentamethyl-4-piperidiny1) (Chimassorb® 119);
- 1,5-dioxaspiro (5,5) undecane 3,3-dicarboxylic acid, bis (2,2,6,6-tetramethyl-4-peridiny1) ester (Cyasorb®
30 UV-500);

- 10 -

- 1,5-dioxaspiro (5,5) undecane 3,3-dicarboxylic acid, bis (1,2,2,6,6-pentamethyl-4-piperidinyl) ester (Cyasorb® UV-516);
 - N-2,2,6,6-tetramethyl-4-piperidinyl-N-amino-oxamide;
 - 5 - 4-acryloyloxy-1,2,2,6,6-pentamethyl-4-piperidine.
 - 1,5,8,12-tetrakis[2',4'-bis(1",2",2",6",6"-pentamethyl-4"-piperidinyl(butyl)amino)-1',3',5'-triazine-6'-yl]-1,5,8,12-tetraazadodecane.
 - HALS PB-41 (Clariant Huningue S.A.)
 - 10 Nylostab® S-EED (Clariant Huningue S.A.)
 - 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)-pyrrolidin-2,5-dione
 - Uvasorb® HA88
 - 1,1'-(1,2-ethane-di-yl)-bis-(3,3',5,5'-tetra-methyl-
 - 15 piperazinone) (Good-rite® 3034)
 - 1,1',1''-(1,3,5-triazine-2,4,6-triyltris((cyclohexylimino)-2,1-ethanediyl)tris(3,3,5,5-tetramethylpiperazinone); (Good-rite® 3150)
 - 1,1',1''-(1,3,5-triazine-2,4,6-
 - 20 triyltris((cyclohexylimino)-2,1-ethanediyl)tris(3,3,4,5,5-tetramethylpiperazinone) (Good-rite® 3159)
- Preferably, use is made of the oligomer of
- N-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-piperidinol
 - 25 and succinic acid (Tinuvin® 622); poly((6-morpholine-S-triazine-2,4-diyl)(2,2,6,6-tetramethyl-4-piperidinyl)-iminohexamethylene-(2,2,6,6-tetramethyl-4-piperidinyl)-imino) (Cyasorb® UV 3346); polymethylpropyl-3-oxy-
 - [4(2,2,6,6-tetramethyl)-piperidinyl]-siloxane (Uvasil®
 - 30 299); oligomer of 7-oxa-3,20-

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diazadispiro[5.1.11.2]heneicosan-21-one, 2,2,4,4-tetramethyl-20-(oxiranylmethyl)- (Hostavin® N30);
 propanedioic acid, [(4-methoxyphenyl)methylene]-bis(1,2,2,6,6-pentamethyl-4-piperidinyl) ester
 5 (Sanduvor® PR 31); 1,3,5-triazine-2,4,6-triamine, N,N'''-[1,2-ethanediylbis [[4,6-bis[butyl(1,2,2,6,6-pentamethyl-4-piperidinyl)amino]-1,3,5-triazine-2-yl]imino]-3,1-propanediyl]]-bis[N',N''-dibutyl-N',N'''-bis(1,2,2,6,6-pentamethyl-4-piperidinyl) (Chimassorb®
 10 119); HALS PB-41 (Clariant Huningue S.A.); Uvasorb® HA88; 1,1',1'''-(1,3,5-triazine-2,4,6-triyltris ((cyclohexylimino)-2,1-ethanediyl)tris(3,3,5,5-tetramethylpiperazinone), (Good-rite® 3150); 1,1',1'''-(1,3,5-triazine-2,4,6-triyltris ((cyclohexylimino)-2,1-ethanediyl)tris(3,3,4,5,5-tetramethylpiperazinone)
 15 (Good-rite® 3159).

In principle, the HALS content of the composition may vary within a broad range, for example between 0.05 and 3 wt.%, preferably between 0.1 and 2
 20 wt.%, even more preferably between 0.2 and 1 wt.%. When HALS without activated carbon is used, the upper limit of the amount of to be used is determined to an important extent by the HALS bleeding effect, but it has, very surprisingly, been found that this effect is
 25 less prominent in the presence of activated carbon. Preferably, high-molecular HALS is used in polyetherester film, for example Tinuvin® 622, Uvasil® 299, Cyasorb® UV 3346 and Chimassorb® 944, which are oligomeric or polymeric HALS compounds and have a
 30 molecular weight > 1500, preferably > 2000.

In practice, therefore, the HALS content of the composition according to the invention is

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determined predominantly by the concentration needed for achieving the desired life. This content generally lies between 0.1 and 1.0 wt.%.

The composition may optionally comprise
5 other additives, for example fillers, thermooxidative stabilizers, further supplementary light stabilizers, for example triazoles or thioesters, processing aids, colourants, etc.

The composition can be obtained in a manner
10 known to the average one skilled in the art, who will pay attention in particular to a good dispersion of the activated carbon and HALS in the composition, preferably using the "masterbatch" method, in which preferably a concentrate of the activated carbon (for
15 example 10-40 wt.%) in the polyether ester or a compatible polymer is produced on a twin-screw extruder, which is then incorporated as required in the melt in the polyetherester copolymer. For this, too, preferably a twin-screw extruder is used. The HALS is
20 also preferably added in the form of a masterbatch.

It is, for that matter, also possible to incorporate the constituent components into the melt in any other way, in doing which special attention should be paid to obtaining a good dispersion, inter alia
25 through almost complete disintegration of the carbon agglomerates in the primary aggregates, by means of inter alia high shear forces or the use of dispersants.

The invention will now be elucidated on the basis of the following examples and comparative
30 experiments. The examples are limited to one type of copolyetherester and a specific combination of carbon black and HALS. However, it will be evident that, as the mechanism of the degradation of the copolyetherester affects the ether groups of the

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copolyether, any other copolyetherester could have been used as a model and the activity of the activated carbon resides in its large specific optical surface area and the activity of the HALS is determined mainly
5 by the presence of the group outlined in the formulas, any other activated carbon types and other HALS will lead to a comparable result.

Materials used:

- 10 Arnitel[®] EM 400: a copolyetherester with polybutylene terephthalate as hard segment and tetramethylene oxide as soft segment, Shore D = 40, from DSM, The Netherlands.
- 15 Activated carbon: (CB) Black Pearls 880 masterbatch, in PE as carrier, from Cabot, average particle size 16 nm.
- HALS: Chimasorb[®] 944, an oligomer of cyanuric acid and N,N-Di(2,2,6,6-tetramethyl-4-
20 piperidyl)-hexamethylene-diamine from Ciba-Geigy, Switzerland.

Experimental:

- 25 The compositions given in Tables 1 and 2 were obtained by blending the Arnitel[®] EM400, the activated carbon and the HALS on a Werner & Pfleider 30 mm twin-screw extruder at a temperature setting of 245°C, a speed of 250 rpm and a throughput of 10 kg/hour. All raw
30 materials were fed to the throat. The granules were processed into a film by means of a Battenfeld 45 mm single-screw extruder, equipped with a Verbruggen 250 mm wide film head. The temperature setting was 220°C

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and the lip was set at 0.3 mm. Variation in the film thickness was obtained through variation of the take-off rate.

5 Ageing:

Ageing was effected by exposing film samples in an Atlas WOM Ci 65A (weather-o-meter) apparatus equipped with Xenon light source under the following conditions:

	Filters	internal	: borosilicate "S"
10		external	: borosilicate "S"
	Intensity		: 0.35 W/m ² /nm at 340 nm
	Temperature:	Black standard	: 67°C
		Black panel	: 73°C
		Area	: 45°C
15	Relative humidity		: 50 ± 10%
	Dry/wet cycle		: 102/18 minutes

At regular intervals samples were taken and subjected to mechanical tests.

20

Mechanical testing:

The elongation at break of the films was determined on a Zwick 1445 tensile tester according to ASTM G 26.

Test bar dimensions according to DIN 53504S3, prismatic
25 part, length: 12 mm, width: 2 mm.

Composition and results:

The life of the samples was characterized as the period after which the elongation at break has decreased to a
30 value below 100%.

Table 1

Film thickness 100 μ m						
Experiment	1	2	3	4	5*	6*
HALS, wt. %	0	1	0	0	1	0.5
CB, wt. %	0	0	1	2	2	1
life [hours]	24	200	150	600	7000	2000
Film thickness 175 μ m						
Experiment	7	8	9*	10*		
HALS, wt. %	0.5	0	1	0.5		
CB, wt. %	0	1	1	2		
life [hours]	168	150	>7000	>7000		
Film thickness 25 μ m						
Experiment	11	12	13*	14*	15	
HALS, wt. %	0	0.5	1	0.5	0	
CB, wt. %	1	0	1	2	0	
life [hours]	20	48	168	150	10	

*) Examples according to the invention.

- 5 The above table shows, as is to be expected, that the life of very thin film is substantially shorter than that of thicker film. The activity of CB (activated carbon) and HALS, both taken separately, also seems to depend on the thickness.
- 10 Especially in the 25 μ m film the activity of CB is inferior. The synergistic effect of the combination of CB + HALS is surprisingly great.

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At a film thickness of 100 μm the life is found to increase almost tenfold compared to the sum of the separate effects of HALS and CB.

5 Experiment 16

In a further experiment a 50 μm film of Arnitel[®] PM 380^{*)} with 1% HALS and 1% CB was tested for its ageing properties under the same conditions as in the preceding experiments. After 1000 hours an
10 elongation at break of 300% is still measured. This again proves the stability of the composition according to the invention.

15 ^{*)}Arnitel[®] PM 380: a polyetherester copolymer with ethylene oxide-capped polypropylene oxide as soft segment. Shore D = 38, from DSM, The Netherlands.

Experiments 17-24

Some Arnitel[®] EM 400 films (100 μm)
20 containing different combinations of UV stabilizers were subjected to ageing tests. The test conditions differed from those of the preceding tests. The intensity was 0.28 W/m²/nm at 340 nm and the black panel temperature 50°C. As a measure of the life again the
25 time was used after which the elongation at break has decreased to less than 100%.

Table 2

Experiment	17	18	19	20	21	22	23	24*
HALS Tin 144	0.5		0.25	0.50	0.0			0.5
UV stab Tin 234		0.5	0.25	0.50	0.0			
CB					0.0	0.4	1.0	1.0
life [hours]	125	400	280	600	48	80	250	1500

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Tin 144 = Tinuvin® 144, M = 665

Tin 234 = Tinuvin® 234, a UV absorber, 2(2-hydroxy-3,5
di(1,1-dimethyl benzyl/phenyl)-
5 2H-benzotriazol

N.B. In the composition without CB the presence of an
antioxidant was necessary; in this case this was 0.25
wt.% Irganox®

1010, a phenolic antioxidant from Ciba.

10

This experiment again proves the highly
reinforcing effect of activated carbon and the HALS
stabilizer. An added advantage of the combination
according to the invention, experiment 24, is that no
15 antioxidant needs to be added.

C L A I M S

1. Polyetherester copolymer composition containing
0.1-10 wt.% activated carbon and 0.05-3 wt.% of a
5 hindered amine light stabilizer (HALS).
2. Polyetherester copolymer composition according to
claim 1, characterized in that the composition
contains 0.5-5 wt.% activated carbon and 0.1-2
wt.% HALS, preferably 0.5-3 and 0.2-1 wt.%,
10 respectively.
3. Polyetherester copolymer composition according to
claim 1 or 2, characterized in that the ratio
between hard and soft segments in the
copolyetherester results in a Shore D hardness of
15 25-80.
4. Polyetherester copolymer composition according to
any one of the preceding claims, characterized in
that the C:O ratio in the polyalkylene oxide
glycol, from which the polyetherester copolymer
20 is derived, is between 2 and 4.3.
5. Polyetherester copolymer composition according to
any one of the preceding claims, characterized in
that the average primary particle size in the
carbon dispersion in the composition is 15-40 nm.
- 25 6. Polyetherester copolymer composition according to
any one of the preceding claims, characterized in
that the HALS has a molecular weight higher than
1500, preferably higher than 2000.
7. Polyetherester copolymer composition according to
claim 6, characterized in that the HALS is chosen
30 from the group formed by Tinuvin® 622, Uvasil®
299, Cyasorb UV 3346 and Chimassorb® 944.
8. Film obtained from a composition according to any
one of claims 1-7.

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9. Film obtained from a polyetherester copolymer composition that contains 0.5-3 wt.% activated carbon and 0.1-2 wt.% HALS, that has a Shore hardness of 25-80, and of which the C:O ratio in the polyalkylene oxide glycol, from which the polyether ester is derived, is between 2 and 4.3, the average particle size in the carbon dispersion in the composition is 15-40 nm and the HALS has a molecular weight higher than 2000.
10. Use of the film according to claim 8 or 9 in building applications.
11. Use of the film according to claim 8 or 9 in roof structures.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/NL 99/00331

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08K13/02 C08K5/00 C08L67/02 //(C08K5/00,5:3435,5:3462,
5:353,5:3492),(C08K13/02,3:04,5:00)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	EP 0 885 924 A (HOECHST CELANESE) 23 December 1998 (1998-12-23) page 7, line 14; claims 1,6; examples page 14, line 18 - line 19 ---	1-3,5,6
A	EP 0 783 016 A (AKZO NOBEL) 9 July 1997 (1997-07-09) cited in the application page 2, column 1, line 32; claims 1,5 ---	1,5
A	WO 96 28503 A (NYLTECH FR) 19 September 1996 (1996-09-19) page 1, line 17 - page 2, line 24; claims 1,2,7 ---	1,2
A	US 4 524 165 A (MUSSEY ET AL.) 18 June 1985 (1985-06-18) claims 1,9,25; tables ---	1,7
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Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
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"A" document member of the same patent family

Date of the actual completion of the international search

6 September 1999

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/NL 99/00331

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A,P	WO 98 39503 A (DU PONT) 11 September 1998 (1998-09-11) page 3, paragraph 1 - paragraph 2; claim 1 -----	1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/IL 99/00331

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WO 9839503 A	11-09-1998	NONE	